

8483M

(19)



Europäisches Patentamt

European Patent Office

Office européen des brevets



(11)

EP 0 841 362 A2

(12)

EUROPEAN PATENT APPLICATION

(43) Date of publication:
13.05.1998 Bulletin 1998/20

(51) Int Cl.⁶: C08J 3/12, C08J 3/16,
B01D 17/06

(21) Application number: 97308866.9

(22) Date of filing: 05.11.1997

(84) Designated Contracting States:
AT BE CH DE DK ES FI FR GB GR IE IT LI LU MC
NL PT SE
Designated Extension States:
AL LT LV MK RO SI

- Hall, Charles Alan
Madison, Indiana 47250 (US)
- Halm, Roland Lee
Midland, Michigan 48640 (US)

(30) Priority: 12.11.1996 US 747531

(71) Applicant: DOW CORNING CORPORATION
Midland Michigan 48686-0994 (US)

(74) Representative: Kyle, Diana
Elkington and Fife
Prospect House
8 Pembroke Road
Sevenoaks, Kent TN13 1XR (GB)

(72) Inventors:
• Gatti, Christopher Darren
Hebron, Kentucky 41048 (US)

(54) Electric field enhanced coalescence of silicone emulsions

(57) A process for separating an emulsion into separate and easily recoverable phases is disclosed. The process comprises exposing said emulsion, comprising a discontinuous phase and a non-conducting continuous phase, to an electric field thereby effecting coalescence of said discontinuous phase into droplets of a size for effective gravitation from the continuous phase,

where the discontinuous phase and the continuous phase have different dielectric constants and densities and at least one of the phases comprise a silicon containing compound or a silicon containing polymer. This process is especially useful for separating emulsions where the discontinuous phase is an aqueous acid solution and the continuous phase is diorganopolysiloxane.

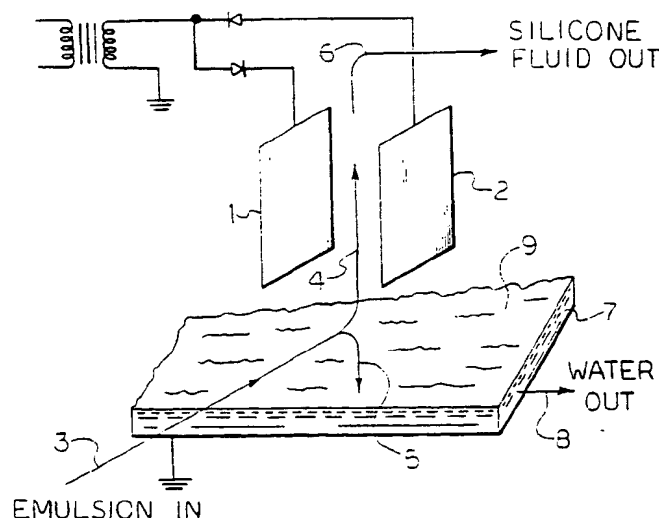


Fig. 1

EP 0 841 362 A2

Description

The present invention is a process for separating an emulsion by electric field enhanced coalescence, where the emulsion comprises a discontinuous phase and a non-conducting continuous phase and the two phases have different dielectric constants and densities and at least one of the phases comprises a silicon containing compound or a silicon containing polymer. Herein, the emulsion is exposed to an electric field thereby effecting coalescence of the discontinuous phase into droplets of a size for effective gravitation from the continuous phase.

The use of high voltage electric fields to force the separation of oil field emulsions is a well known and accepted practice in the petroleum industry. These fields greatly speed the coalescence and separation of immiscible liquids, over conventional heat treaters and settlers using mechanical aids to accomplish coalescence.

This art is generally represented by U.S. Patents 3,207,686 and 3,342,720.

Moreover, in processes for dehydrating crude petroleum-aqueous emulsions by effecting coalescence in an electric field, the nature of the current provided to the electrodes is important in determining process efficiency. It is believed that an electric field created by an AC voltage source is more effective in separating relatively wet emulsions while an electric field created by a DC voltage source is more effective in separating dryer emulsions where particle size of the aqueous phase is small. Therefore apparatuses employing electrical fields of both AC and DC characteristics have been described.

This latter art is represented by U.S. Patents 3,772,180; 3,939,395; 4,054,451; 4,308,127 and 4,126,537.

The above described patents specifically address the problems associated with dehydrating crude oil and petroleum distillates in the petroleum industry. Said patents do not recognize that silicon containing compounds and silicon containing polymers represent unique materials that can exist in emulsion with other liquids and that such emulsions with two phases having different dielectric constants and densities may be separated by exposure to an electric field to enhance coalescence of the discontinuous phase into droplets of a size for effective gravitation from the continuous phase.

The invention is a process for separating an emulsion into separate and easily recoverable phases. The process comprises exposing an emulsion comprising a discontinuous phase and a non-conducting continuous phase to an electric field thereby effecting coalescence of the discontinuous phase into droplets of a size for effective gravitation from the continuous phase, where the discontinuous phase and the continuous phase have different dielectric constants and densities and at least one of the phases comprises a silicon containing compound or a silicon containing polymer. Our process is especially useful for separating emulsions where the discontinuous phase is an aqueous acid solution and the continuous phase is diorganopolysiloxane.

Figure 1 is a schematic overview of an embodiment of the present process as it relates to the separation of an emulsion of water in a polydiorganosiloxane polymer.

Figure 2 is a schematic representation of significant elements of an apparatus suitable for conduct of the present process.

Our method of exposing the emulsion to an electric field and the apparatus for effecting such exposure is not critical to the present invention and can be any of those known in the art. By way of example, an overview of an embodiment of the present process where the continuous phase is a silicone fluid, such as polydimethylsiloxane, and the discontinuous phase is water is presented in Figure 1. In Figure 1, electrodes 1 and 2 are shown, arranged vertically and parallel to each other and to the flow of fluids within a treater shell. The emulsion is directed toward the electrodes along path 3 which is generally lower than the electrodes. The flow of the emulsion from path 3 is then directed upward along path 4. Any water droplets that coalesce to a size large enough to gravitate from the emulsion travel downward along path 5. After treatment by exposure to electric fields, the silicone fluid continues upward along path 6 for delivery as a dehydrated product of the process.

The water which is separated from the emulsion flowing along path 5, collects as bulk water 7. A controlled removal of water from bulk water 7 is effected along path 8. The controlled removal of water is exerted to maintain water interface 9 at a predetermined distance below electrodes 1 and 2. In Figure 1, the potential applied to the electrodes is shown as a DC voltage generated by rectifying AC voltage into half-wave negative and positive components thereby providing a pulsed positive DC voltage to one electrode and a pulsed DC negative voltage to the other electrode.

The nature of the electrical potential supplied to the electrodes in Figure 1 is not meant to be limiting on the present process and this potential can be supplied as an AC potential, DC potential, pulsed DC potential, a combination of AC and DC potentials or as an electrical potential having both AC and DC characteristics. One preferred process of our invention uses one or more pairs of electrodes connected to an AC energized transformer, one end of the transformer secondary winding being connected to ground and the other connected to the electrodes through rectifier elements, such that the AC supply is split into its pulsed unidirectional components. On each half cycle of the AC voltage source, voltage is applied to one electrode of the pair while the other electrode of said pair has no voltage applied to it. A fluctuating unidirectional field is therefore produced between the electrodes by the alternate pulsing of one electrode positive and the other electrode negative. Under conditions of Figure 1, the pulsating DC voltage applied to the elec-

trodes may establish a field of an AC nature between the ends of the electrodes and the water interface 9 which is maintained at ground potential.

The method of exposure of the emulsion to the electric field and an apparatus suitable for such exposure is further detailed by Figure 2. In Figure 2, electrodes 1 and 2 are shown arranged vertically and parallel to each other and to the flow of fluids within treater shell 10. Electrodes 1 and 2 are connected to power supply 11 by means of electrical leads 12 and 13. Power supply 11 is a rectified AC voltage providing a pulsating DC voltage to the electrodes. Also positioned within treater shell 10 is distributor 14. Distributor 14 is of conventional design and comprises, for example, a notched or perforated inverted conduit having an H-shaped configuration. The emulsion is conveniently fed to distributor 14 along path 3 consisting of feed conduit 15 having positioned therein a feed control valve 19 and a flow meter 20. Feed control valve 19 is used to control the rate of feed of emulsion to distributor 14. The emulsion is preferably fed into treater shell 10 at a level below distributor 14 and above water interface 9.

Water interface 9 is kept at a relatively constant level below distributor 14 by removal of bulk water 7 from treater shell 10 through outlet 16. The exit of bulk water 7 from the treater shell is controlled by means of outlet valve 17 which may be connected to an automatic level control system such as a float valve arrangement. The emulsion exiting distributor 14 is channeled by channeling plates 21 along path 4 passing between electrodes 1 and 2. The purpose of channeling plates 21 is to ensure the emulsion passes along path 4 and not behind electrodes 1 and 2. Channeling plates 21 are made of an inert non-conductive material such as Teflon (R) (Dupont Corporation, Wilmington, Delaware). As the emulsion passes between electrodes 1 and 2, coalescence of the aqueous phase is effected causing the formation of drops large enough to settle by gravitation from the emulsion. The aqueous drops settle into bulk water 7. The dehydrated silicone fluid continues upward along path 6 through treater shell 10 exiting at fluid outlet 22 and passes through conduit 23 to storage container 24.

It will be readily appreciated by those skilled in the art that the embodiments of the present invention as described by reference to Figure 1 and Figure 2 are not exclusive and that other embodiments could be readily fashioned from the disclosure herein without the exercise of inventive skill. More, specifically the number, configuration, design and location of the electrodes; the type of voltage supplied to said electrodes, including the method of control; the distributor, number, design and location; the methods for feeding dispersion to the electric field coalescer unit; and methods for removing separated components from the coalescer can be any of those typically associated with such processes. Specific examples of electric field coalescer units which are useful in our process include those described in U.S. Patents 3,342,720; 3,207,686; 3,772,180; 4,056,451 and 4,126,537.

Emulsions that are readily separated by the present process comprise at least two immiscible liquids where one of the liquids is non-conducting and is present as a continuous phase. By "non-conducting", we mean the liquid comprising the continuous phase has a conductivity such that the dielectric resistance between energized electrodes is maintained high enough to sustain an electric field between the electrodes sufficient to effect coalescence of the discontinuous phase.

The nature of the continuous phase is limiting only in that under process conditions the continuous phase will be non-conductive and have a dielectric constant and density different than that of the discontinuous phase. The continuous phase is, for example, an organic solvent such as toluene, xylene, benzene and heptane.

Preferred is when the continuous phase comprises a silicon containing compound or a silicon containing polymer. By silicon containing compounds, we mean those chemical entities comprising at least one silicon atom and being non-polymeric in structure. Examples of silicon containing compounds which comprise the continuous phase of an emulsion separable by the present process include silanes described by formula



where each R^1 is an independently selected monovalent hydrocarbon radical comprising 1 to 12 carbon atoms. X is a halogen atom, $a=0$ to 4, $b=0$ to 2 and $a+b=0$ to 4. Each R^1 is selected, for example, from a group consisting of substituted and unsubstituted alkyls, cycloalkyls, alkenyls, aralkyls and aryls. R^1 is, for example, methyl, ethyl, chloromethyl, 3,3,3-trifluoropropyl, cyclopentyl, cyclohexyl, vinyl, allyl, 5-hexenyl, benzyl, beta-phenylethyl, phenyl, tolyl, xylyl, naphthyl and chlorophenyl. Preferred is when R^1 is methyl. In Formula 1, each X is an independently selected halogen atom. Preferred in Formula 1 is where X is chlorine.

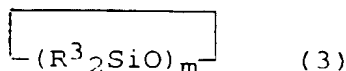
The continuous phase of an emulsion separable by the present process can also be a silicon containing polymer. By silicon containing polymer, we mean those polymers in which the silicon atom is a substituent of the repeating units forming the polymer backbone chain. Examples of such silicon containing polymers include linear, branched and cyclic siloxanes; silicone resin precursors; polysilanes; and mixtures of two or more of these polymers.

The silicon containing polymer is, for example, a linear siloxane described by formula



where each R^2 is independently selected from the group consisting of halogen, hydroxy and R^3 ; each R^3 is independently selected from hydrogen or monovalent hydrocarbon radicals comprising 1 to 12 carbon atoms and z is a value within a range of 0 to 100,000. In formula (2), it is preferred that each R^2 is independently selected from the group consisting of chlorine, hydroxy and methyl. Also, in formula (2), R^3 is in addition to hydrogen, a substituted or unsubstituted alkyl, cycloalkyl, alkenyl and aryl as described for R^1 . Preferred is where at least 50 mole percent of the R^3 substituents on silicon in formula (2) are methyl. It is also preferred in formula (2) that z be a value within a range of 0 to 700. More preferred is when z is within a range of 0 to 200.

The silicon containing polymer is, for example, a cyclic siloxane described by formula



where R^3 is as previously described and m is a value within a range of 3 to 20. Preferred is where m is within a range of 3 to 7.

The silicon containing polymer is typically a branched polymer, suitable as a precursor for forming a silicone resin of units described by formulas $R^3_3SiO_{1/2}$, $R^3_2SiO_{2/2}$, $R^3SiO_{3/2}$ and $SiO_{4/2}$, where R^3 is as previously described.

The silicon containing polymer can also be, for example, a polysilane described by formula



where each R^4 is independently selected from the group consisting of hydrogen, halogen, R^1 as previously described and n is a value within a range of 0 to 1,000.

The present process is particularly useful for reducing residual water and chloride present in polysiloxanes prepared by the hydrolysis of organochlorosilanes. Such a process is described, for example, in U.S. Patent 5,075,479, which teaches emulsions comprising silicon containing polymers as a continuous phase and an aqueous acid as a discontinuous phase that are separated by the present process.

The discontinuous phase of the present process is any liquid immiscible with the continuous phase and having a dielectric constant and density different than that of said continuous phase. The discontinuous phase can be an organic solvent, silicon containing compound or silicon containing polymer such as described for the continuous phase; water; or aqueous acid. Within the limitations described for the continuous phase and the discontinuous phase, our claimed process is used to remove residual amount of silicon containing compounds and silicon containing polymers from solvents to facilitate recovery, recycle or disposal of solvents. In a preferred process, the discontinuous phase of the emulsion is an aqueous acid, such as results from the hydrolysis of organohalosilanes to form organopolysiloxanes. The discontinuous phase is, for example, aqueous hydrogen chloride.

The discontinuous phase of the emulsion must have a different dielectric constant than that of the continuous phase. The required difference in dielectric constant is dependent upon the particular configuration of electric field coalescer used to effect the present process and can be easily determined without undue experimentation by those skilled in the art. Generally, the dielectric constant of the discontinuous phase must be sufficiently different from that of the continuous phase to allow for migration of the droplets forming the discontinuous phase within the electric field created between charged electrodes. The discontinuous phase can also be electrically conductive. Examples of materials having differing dielectric constants that can be separated by the present process are provided herein.

The material comprising the discontinuous phase must also have a different density than that of the material comprising the continuous phase. The present process effects coalescence of the discontinuous phase into droplets of a size for effective gravitation from the continuous phase. Therefore, the density of the material comprising the discontinuous phase must be such that coalesced droplets of the discontinuous phase are separated from the continuous phase based upon their buoyancy or settling in the continuous phase. In the present process, the coalesced droplets of the discontinuous phase have a density that allows them to either migrate up or down within the continuous phase to effect a phase separation. In a preferred process the discontinuous phase has a density greater than that of the continuous phase causing the coalesced droplets of the discontinuous phase to settle toward the bottom of an electric field coalescer apparatus.

When the continuous phase is a silicon containing polymer, it may be desirable herein to add water to our process

to facilitate reduction of residual chlorine content of the silicon polymer. The added water, comprising at least a portion of the discontinuous phase of the emulsion, will facilitate hydrolysis of chlorine atoms bonded to silicon atoms, thereby facilitating removal of chlorine from said silicon containing polymer.

The following examples are provided to further illustrate the present invention.

Example 1

The ability to dehydrate and reduce the chlorine content of a polydimethylsiloxane (PDMS) fluid by electric field enhanced coalescence was evaluated. Aqueous hydrogen chloride solution having the acid concentration described in Table 1 (Inlet HCl) was emulsified in a PDMS fluid having a viscosity of 0.1 Pa·s at 25°C. The volume percent water (Inlet %H₂O) and parts per million chloride (PPM Cl) in the emulsion is described in Table 1 for each run. The particle size of the aqueous acid phase in the emulsion was determined to be 20 micrometers or less with a median particle size of 10 as measured by use of a particle size analyzer (Lasentec Sensor Technology, Inc., Bellevue, WA). The emulsion was fed to an electric coalescer as generally described in Figure 2. The volume capacity of the coalescer was 64.4 l. (17 gallons). The coalescer was supplied with a rectified voltage of 18 kV and the emulsion was fed to the coalescer at a rate of 1200 ml/m. It was determined that the coalescer reached a steady state condition after 2.5 vessel displacements and a sample of the PDMS exiting the coalescer was collected and analyzed at this point for volume percent water (Outlet %H₂O) and parts per million (PPM) chloride (Outlet PPM Cl). The volume percent water in the outlet stream was determined by standard methods using centrifugation to effect separation. The PPM Cl in the outlet stream was determined by ion chromatography (IC). The results of these analyses are reported in Table 1.

Table 1

PDMS Dehydration and Chloride Removal						
Run	Temp. (°C)	Inlet HCL Conc. (Wt.%)	Inlet		Outlet	
			%H ₂ O	PPM Cl	%H ₂ O	PPM Cl
1	38	1 - 1.1	2.4	250	trace	2.8
2	31	2 - 2.1	2.3	473	trace	0.6
3	24	8.4 - 8.5	9	7594	0.02	2.7

Example 2

The effect of feed velocity on separation of an emulsion of water in PDMS and electric field coalescer was evaluated. An emulsion of water in PDMS fluid having a viscosity of 0.1 Pa·s at 25°C. was created. The water droplet size in said emulsion was similar to that described in Example 1. The emulsion was fed to the electric field coalescer of Example 1 at various feed rates (Feed Rate) as described in Table 2 providing a velocity (Velocity) within the coalescer as also reported in Table 2. The coalescer was run at ambient temperature. The volume percent water emulsified in the PDMS fed to the reactor (Inlet %H₂O) and the volume percent water in the PDMS exiting the reactor (Outlet %H₂O) are also in Table 2. The weight percent water was determined by standard methods using centrifugation to effect separation.

Table 2

Effect of Emulsion Feed Velocity on Dehydration			
Feed Rate (ml/s)	Velocity (X10 ³ m/s)	Inlet %H ₂ O	Outlet %H ₂ O
20	1.29	8	0.03
40	2.59	5.5	0.07
56	3.63	5.5	0.10
80	5.78	6	0.12
160	10.31	5	0.40

Example 3

The effect of the voltage applied to the electric field coalescer on the ability to separate water in a PDMS emulsion was evaluated. An emulsion of water in PDMS fluid having a viscosity of 0.1 Pa·s at 25°C. was created. The volume percent of water emulsified in said fluid is reported in Table 3 (Inlet %H₂O). The water droplet size in the emulsion was similar to that described in Example 1. The emulsion was fed to the electrostatic coalescer of Example 1 at 1200 ml/

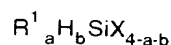
m at various applied voltages as listed in Table 3. The coalescer was run at ambient temperature. Incremental increases in the applied voltage to the coalescer were made as described in Table 3. At each voltage the coalescer was allowed to reach steady state as described in Example 1 before a sample was taken from the exit stream for analysis. The volume percent water emulsified in the PDMS fluid fed to the reactor (Inlet %H₂O) and the volume percent water in the PDMS fluid exiting the reactor (Outlet %H₂O) are reported in Table 3. The volume percent water was determined in the outlet liquid by standard methods using centrifugation to effect separation.

Table 3

Effects of Applied Voltage on Dehydration of PDMS		
Applied Voltage (kV)	Inlet %H ₂ O	Outlet %H ₂ O
18	2.2	0.050
20	2.3	0.035
22	2.5	0.035
24	2.7	0.035
26	2.5	0.035
28	2.6	0.035
30	2.6	0.035
32	2.5	0.030
34	2.6	0.020
36	2.6	0.020
38	2.7	0.020
40	2.7	0.025
42	2.8	0.025
44	2.9	0.025
46	3.0	0.025
48	3.0	0.050

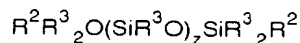
Claims

1. A process for separating an emulsion, the process comprising exposing an emulsion comprising a discontinuous phase and a non-conducting continuous phase to an electric field effecting coalescence of the discontinuous phase into droplets of a size for effective gravitation from the continuous phase, where the continuous phase and the discontinuous phase have different dielectric constants and densities and at least one of the phases comprises a silicon containing compound or silicon containing polymer.
2. A process for dehydrating a silicone polymer, the process comprising passing an emulsion comprising a discontinuous aqueous phase and a continuous phase comprising a polydiorganosiloxane through an electric field having an alternating current field component and a direct current field component, thereby effecting coalescence of the aqueous phase into droplets of a size sufficient for gravitation from the polydiorganosiloxane.
3. A process according to claims 1 or 2 where the electric field is created by an alternating current potential or a direct current potential applied to one or more electrodes.
4. A process according to claims 1 or 2 where the electric field is created by a pulsed direct current potential applied to one or more electrodes.
5. A process according to claims 1 or 2 where the electric field is created by an electric potential having both alternating current and direct current characteristics applied to one or more electrodes.
6. A process according to claim 1 where the continuous phase comprises a silicon containing compound or a silicon containing polymer.
7. A process according to claim 1 where the continuous phase is a silane described by formula



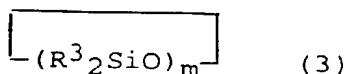
where each R^1 is an independently selected monovalent hydrocarbon radical comprising 1 to 12 carbon atoms. X is a halogen atom, $a=0$ to 4, $b=0$ to 2 and $a+b=0$ to 4.

8. A process according to claim 1 where the continuous phase is a linear siloxane described by formula



where each R^2 is independently selected from the group consisting of halogen, hydroxy and R^3 ; each R^3 is independently selected from hydrogen atom or monovalent hydrocarbon radicals comprising 1 to 12 carbon atoms and z is a value within a range of 0 to 100,000.

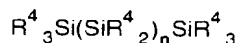
9. A process according to claim 1 where the continuous phase is a cyclic siloxane described by formula



where each R^3 is independently selected from hydrogen atom or monovalent hydrocarbon radicals comprising 1 to 12 carbon atoms and m is a value within a range of 3 to 20.

10. A process according to claim 1 where the continuous phase is a silicone resin formed of units described by formulas $R^3_3 SiO_{1/2}$, $R^3_2 SiO_{2/2}$, $R^3 SiO_{3/2}$ and $SiO_{4/2}$, where each R^3 is independently selected from hydrogen atom or monovalent hydrocarbon radicals comprising 1 to 12 carbon atoms.

11. A process according to claim 1 where the continuous phase is a polysilane described by formula



where each R^4 is independently selected from the group consisting of hydrogen, halogen and monovalent hydrocarbon radicals comprising 1 to 12 carbon atoms and n is a value within a range of 0 to 1,000.

12. A process according to claim 1 where the discontinuous phase is selected from water, aqueous acid and aqueous hydrogen chloride.

13. A process according to claim 2 where the continuous phase is a polydimethylsiloxane prepared by the hydrolysis of methylchlorosilanes, the discontinuous phase is aqueous hydrogen chloride and water is added to the process to facilitate reduction of residual chlorine bonded to silicon atoms of the polydimethylsiloxane.

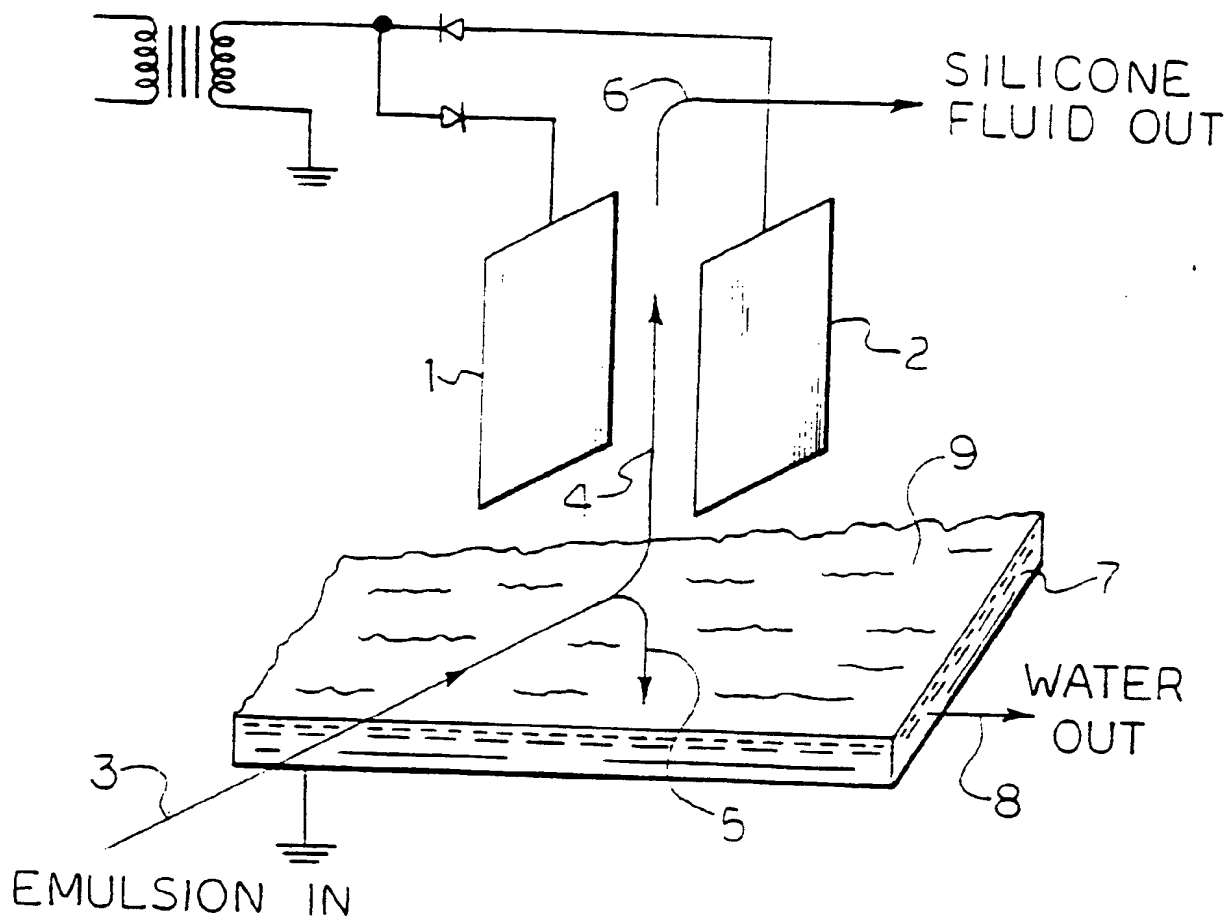


Fig. 1

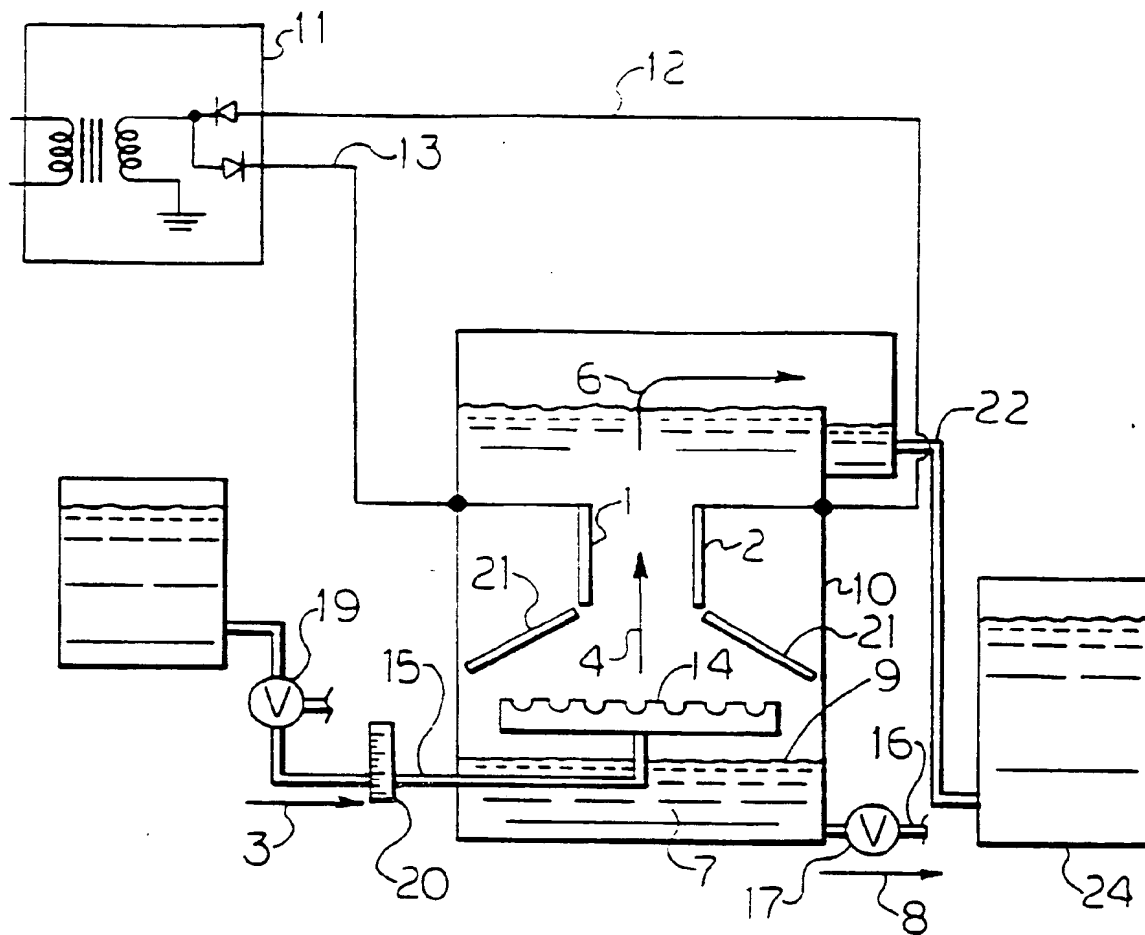
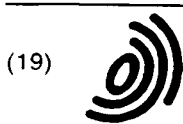


Fig. 2



(19)

Europäisches Patentamt
European Patent Office
Office européen des brevets



(11)

EP 0 841 362 A3

(12)

EUROPEAN PATENT APPLICATION

(88) Date of publication A3:
07.01.1999 Bulletin 1999/01

(51) Int Cl.⁶ C08J 3/12, C08J 3/16,
B01D 17/06

(43) Date of publication A2:
13.05.1998 Bulletin 1998/20

(21) Application number: 97308866.9

(22) Date of filing: 05.11.1997

(84) Designated Contracting States:
AT BE CH DE DK ES FI FR GB GR IE IT LI LU MC
NL PT SE
Designated Extension States:
AL LT LV MK RO SI

- Hall, Charles Alan
Madison, Indiana 47250 (US)
- Halm, Roland Lee
Midland, Michigan 48640 (US)

(30) Priority: 12.11.1996 US 747531

(71) Applicant: DOW CORNING CORPORATION
Midland Michigan 48686-0994 (US)

(74) Representative: Kyle, Diana
Elkington and Fife
Prospect House
8 Pembroke Road
Sevenoaks, Kent TN13 1XR (GB)

(72) Inventors:
• Gatti, Christopher Darren
Hebron, Kentucky 41048 (US)

(54) Electric field enhanced coalescence of silicone emulsions

(57) A process for separating an emulsion into separate and easily recoverable phases is disclosed. The process comprises exposing said emulsion, comprising a discontinuous phase and a non-conducting continuous phase, to an electric field thereby effecting coalescence of said discontinuous phase into droplets of a size for effective gravitation from the continuous phase,

where the discontinuous phase and the continuous phase have different dielectric constants and densities and at least one of the phases comprise a silicon containing compound or a silicon containing polymer. This process is especially useful for separating emulsions where the discontinuous phase is an aqueous acid solution and the continuous phase is diorganopolysiloxane.

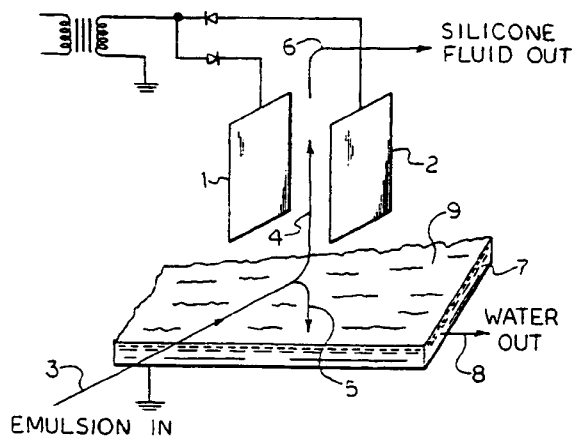


Fig. 1

EP 0 841 362 A3



European Patent
Office

EUROPEAN SEARCH REPORT

Application Number
EP 97 30 8866

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication where appropriate of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
A	WO 90 05589 A (SPECTRUM SCIENCES BV) 31 May 1990 * claims 1-20 *	1	C08J3/12 C08J3/16 B01D17/06
A	WO 88 02274 A (WHITLOCK DAVID R) 7 April 1988 * claims 1-3, 14, 23 * * page 15, paragraph 3 - page 16, paragraph 1 *	1	
A	US 5 075 479 A (BOKERMAN GARY N ET AL) 24 December 1991 * claim 1 * * column 5, line 28 - line 35 *	1	
P.A	DATABASE WPI Derwent Publications Ltd., London, GB; AN 98-310582 XP002083591 & RU 2 094 128 A (ELTRN MATERIALS RES INST), 27 October 1997 * abstract *	1	
The present search report has been drawn up for all claims			TECHNICAL FIELDS SEARCHED (Int.Cl.6)
			B01D B03C C08J
Place of search		Date of completion of the search	Examiner
THE HAGUE		9 November 1998	Depijper, R
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	

EPO FORM 1503 03 82 (F04C11)